AD-A110 848 JOINT INST FOR LAB ASTROPHYSICS BOULDER CO F/G 20/6
OBSERVATION OF ZEEMAN DEGENERACY EFFECTS IN COLLISIONAL INTENSE--ETC(U)
1980 P D KLEIBER, K BURNETT, J COOPER N00014-76-C-0611

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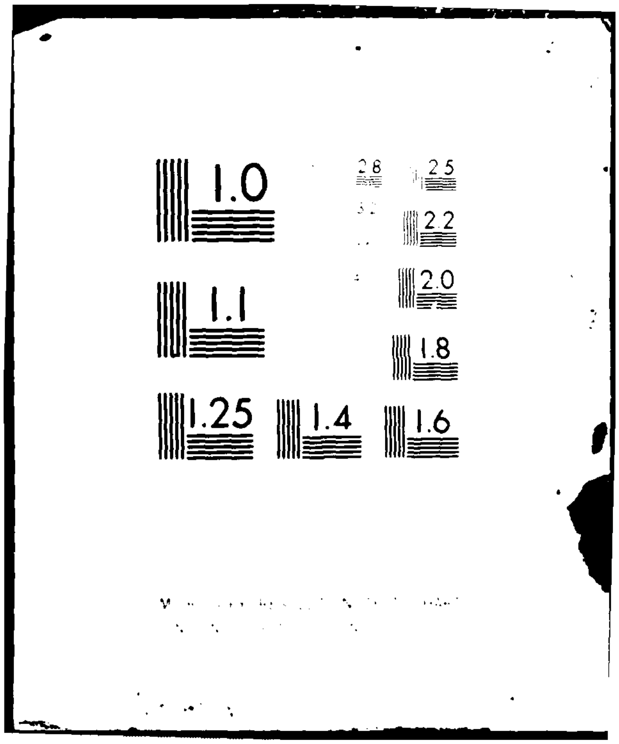
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Observation of Zeeman degeneracy effects in collisional intense field resonance fluorescence

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ABSTRACT

We report observation of the scattered spectrum from a Zeeman degenerate atom at high laser intensities. For a $J = 0$ to $J = 1$ transition in the presence of collisions, we observe an asymmetric triplet in the polarization parallel to the incident laser and an asymmetric doublet in the polarization perpendicular to the incident laser.

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I. Introduction

In recent years there has been widespread interest in high intensity near resonant light scattering in the presence of collisions.[1-6] Mollow[1,2] has developed the theory for a simple two-state atom, including the effects of collisions by introducing phenomenological relaxation terms to the equations of a strongly driven two-state system. This approximation, which is equivalent to the Markoff approximation, leads to equations which are valid in the impact region of the spectrum. The theory predicts the well-known asymmetric triplet structure in the scattered spectrum. These effects have been observed by Carlsten, Szöke and Raymer (CSR)[4] in nonlinear light scattering from strontium perturbed by argon.

Recently, Cooper, Ballagh and Burnett (CBB),[5] and Fiutak and Van Kranendonk[6] have calculated the expected polarized scattering spectrum at high intensities when Zeeman degeneracy effects are included. CBB predict, for the case of a J = 0 to J = 1 transition and a linearly polarized incident laser, an asymmetric triplet scatterd in π polarization, and an asymmetric doublet in σ polarization. The predictions for the π polarized scattering spectrum agree with Ref. 6, however Fiutak and Van Kranendonk do not quote results for the σ polarized spectrum. In this letter we report the observation of these spectral features in the case of strontium perturbed by argon. The qualitative agreement with the results of CBB[5] is quite good.

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II. Theoretical Considerations

The theory for intense field scattering from a Zeeman degenerate atom in a collisional environment has been given by CBB[5] in the impact approximation and steady-state limit. We use the notation of CBB throughout. The impact approximation restricts the validity of their results to pressures low enough such that the duration of a collision (τ_c) is small compared to the time between collisions (T_c). In addition the results are applicable for detunings from line center ($\omega_L - \omega_0 = \Delta$) small compared to the inverse of a collision duration, $\Delta < 1/\tau_c$. For the case of van der Waals broadening $1/\tau_c \sim 10^{12}\ s^{-1}$, thus their results should be valid for detunings $\Delta \lesssim 5\ cm^{-1}$. The theory, furthermore, is valid in the regime where the collision rates are independent of laser intensity, i.e., when $\Omega\tau_c << 1$ where Ω is the Rabi frequency.

Similarly the steady-state assumption restricts their results to laser pulse lengths (τ_p) large compared to the time scales associated with the system relaxation rates ($\tau_p >> T_c, \tau_N$ where τ_N is the radiative lifetime). For buffer gas pressures of ~10 torr and again assuming a van der Waals interaction $T_c \sim \tau_N \sim 5nS$.

For the case of interest here, a linearly polarized laser tuned near the Sr resonance line (a J = 0 to J = 1 transition) and perturbed by Ar (van der Waals broadening) CBB[5] predict a triplet in the scattered spectrum polarized parallel to the incident laser and a [illegible]t in the scattered spectrum polarized perpendicular to the incident laser. In the parallel polarization (Fig. 1a) the three peaks correspond to (i) Rayleigh scattering off the ground or $m_J = 0$ sublevel of the excited state, (ii) "three-photon" scattering followed by fluorescence, and (iii) collision-induced fluorescence from the excited $m_J = 0$ state. These features are exactly those associated with nonlinear scattering from an idealized two-state atom in the presence of collisions.

In the perpendicular polarization the two peaks correspond to collisional excitation of the $m_J = \pm 1$ sublevels of the excited state (Fig. 1b) followed by (i) fluorescence to the ground state or (ii) an inverse Raman excitation of the $m_J = 0$ excited state.

In the experimental results reported here, neither the impact approximation nor the steady-state approximation is strictly valid. Thus detailed quantitative comparisons with the theory are not possible. However, the experimental results are in agreement with the qualitative conclusions of CBB.[5]

III. Experimental Apparatus

The experimental apparatus is basically the same as that used by CSR.[4] The apparatus consists of an N_2 pumped Hänsch type dye laser tuned near the strontium resonance line ($6s^2\ ^1S_0 - 6s6p\ ^1P_1$ at 460.7 nm) and tightly focused into a stainless-steel crossed oven containing strontium vapor at ~500°C-600°C and an argon buffer gas at pressures of ~2 to 500 torr. The laser power at the center of the oven was estimated to be ~1 GW/cm^2. The scattered light was detected at 90° through the side arm of the oven, and focused onto the entrance slit of the monochromator. The signal was detected by a photomultiplier, fed into an rf shielded, gated, analog integrate and hold circuit and recorded on a chart recorder with a 2.2 s time constant. Further details of the apparatus can be found in CSR.[4] The major difference in our experiment was the addition of a glan-prism polarizer on the dye laser output, an analyzing polarizer at the side window output of the oven, and a reference polarizer oriented at 45° and positioned before the entrance slit of the monochromator to remove the polarizing influence of the grating.

IV. Experimental Results

A. Spectrum

Typical spectra are shown in Fig. 2. These spectra were taken at a temperature of ~600°C and argon pressures of 5, 50 and 500 torr respectively. The Sr density, as measured by the curve-of-growth method was ~2×10^{14} cm^{-3} which agreed to ~50% with the vapor pressure curve results. At this density and an Ar pressure of 5 torr the ratio of the van der Waals broadening rate (γ(Sr-Ar)) to the resonance broadening rate (γ(Sr-Sr)) was ~10. The effect of Sr-Sr collisions on the results then should be negligible. As an additional check the experiments were repeated at 550°C (measured Sr density ≈6×10^{13} cm^{-3}) and the results were unchanged. The laser detuning in these spectra was $\Delta = 17$ cm^{-1} (~0.4 nm) to the red. The monochromator resolution was ~0.07 nm and the absolute wavelength calibration is expected to be good to ±0.02 nm.

The triplet and doublet features are clearly visible in the two polarizations at low argon pressures. As the pressure is increased the collisional features dominate over the radiative components (Fig. 2 - 500 torr). Note also that each component (except the Rayleigh peak) has large asymmetric wings. These wings are due to the variation in ac Stark shift that different atoms experience as a result of the spatial intensity dependence of the laser field. The Stark shifts are, of course, away from the Rayleigh peak, hence the asymmetry. In the Stark wings (Stark shift >0.1 nm) the fluorescence path is optically thin, however, the fluorescence around line center is heavily trapped.

The majority of the measurements were done at a laser detuning of 17 cm^{-1} to the red of the Sr resonance line. Changing the detuning had little effect on the qualitative features shown in Fig. 2. With the laser tuned to the blue

of the Sr resonance line the scattered spectrum was again qualitatively similar. However, as expected we found the collisional features to be much weaker for blue detunings, corresponding to absorption in the anti-static wing of the line and subsequent re-emission.

V. Conclusions

We have demonstrated that the experimental techniques of CSR[4] for the study of collisional redistribution and saturation of near resonant scattered light can be easily extended to include polarized scattering experiments. These experiments can obviously give much more information about collision processes in "real" (i.e., Zeeman degenerate) atomic systems including information about m_J-changing collision rates.

We have also verified the qualitative predictions of CBB[5] concerning high intensity near-resonant scattering from a Zeeman degenerate atom in the presence of collisions.

This work was supported in part by the National Science Foundation under Grant No. PHY79-04928 and in part by the Office of Naval Research under Grant No. DOD N00014-76-C0611.

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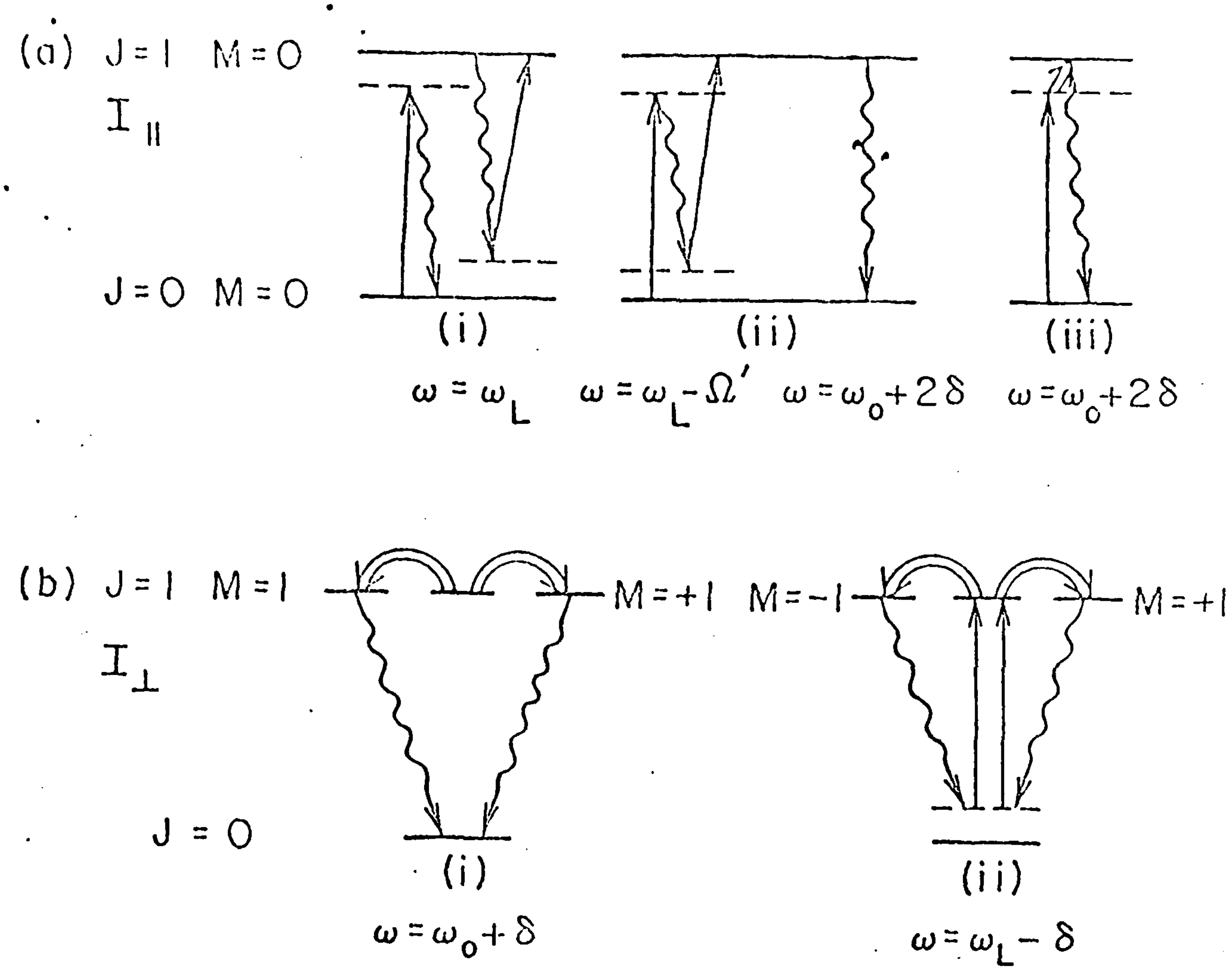
Figure Captions

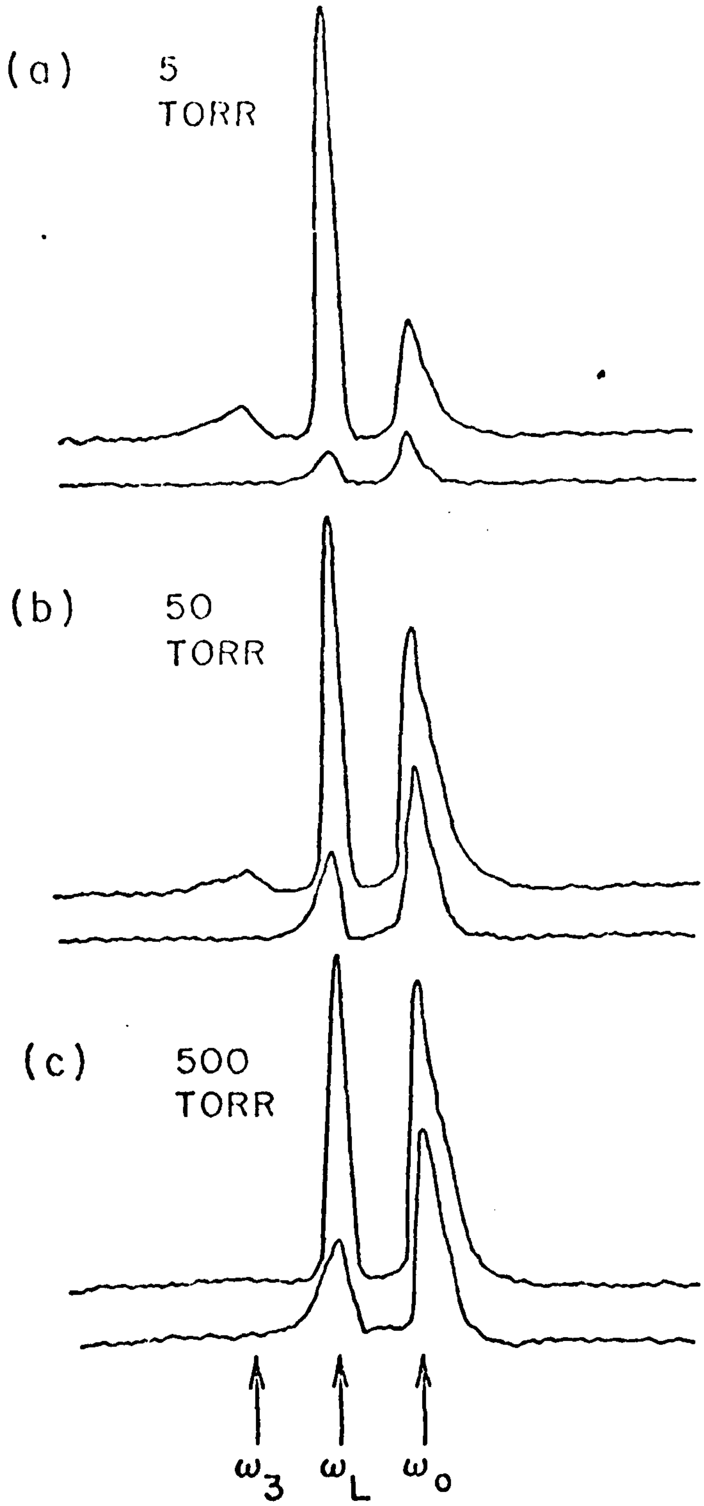
Fig. 1. Processes involved in formation of spectrum (the straight arrows represent absorption of a laser photon, the wavy arrows represent an emitted photon, and the double arrows represent a collisional process)..

Fig. 2. Typical scattered light spectra (red detuning), at (a) 5 torr, (b) 50 torr and, (c) 500 torr. In each case the upper curve is the parallel polarization and the lower curve the perpendicular polarization. The arrows indicate the unshifted positions of the peaks.



(a) J = 1 M = 0
$I_{\parallel}$
J = 0 M = 0
(i)
(ii)
(iii)
$\omega = \omega_L$
$\omega = \omega_L - \Omega'$
$\omega = \omega_o + 2\delta$
$\omega = \omega_o + 2\delta$
(b) J = 1 M = 1
$I_{\perp}$
M = +1
M = -1
M = +1
J = 0
(i)
(ii)
$\omega = \omega_o + \delta$
$\omega = \omega_L - \delta$

Figure 1



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